Persistent Organohalogen Contaminants in Fish from Lakes and Reservoirs in Southwestern British Columbia, Canada

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Abstract
Despite bans or highly restricted registration of persistent organohalogen pesticides and PCBs, these contaminants continue to be detected in the environment. Concern about the result of atmospheric transport and "cold condensation" into both high latitude and high elevation ecosystems has been mounting. Such a mechanism has produced surprising and toxicologically significant concentrations of these chemicals in "pristine" ecosystems. Westerly flows from potential source areas in Asia and atmospheric release from the North Pacific coupled with the "trapping" effect of high western mountains present a perfect situation for organohalogen accumulation. This survey explored the extent and significance of POPs residues in south west British Columbia through the analysis of selected organochlorine compounds in: (1) fish tissues from remote lakes and reservoirs and, (2) high-elevation snowpack.

Dominant salmonid species were sampled from a total of 17 lakes and reservoirs covering a wide range of physiographic conditions (elevation (0-1435m), watershed basin area/character, bathymetry) in SW BC. Skin-off muscle fillets were analyzed for a range of non-ortho PCB congeners, organochlorine pesticides and toxaphene. Snow samples were collected from a total of 9 high elevation (1200 – 2450m) sites in the mountains surrounding the Strait of Georgia between Vancouver Island and mainland British Columbia. The snow was melted (68-75L total volume), extracted with XAD-2 resin, and analyzed for a suite of PCB congeners, OC pesticides and toxaphene.

Introduction
Studies of organochlorine pollutant residues in fish, birds, mammals and snow from remote Arctic and alpine sites in Canada and elsewhere have shown the potential for surprisingly high concentrations of atmospherically transported contaminants (Campbell et al. 2000, Donald et al. 1993, Grimalt et al. 2001, Kidd et al. 1998). Sites are often distant of potential sources, and evidence is increasing that emissions throughout the northern hemisphere are contributing to contaminant loading to these areas (Bailey et al. 2000, Wania 1999, Wania and Mackay 1996). For example, there is evidence for trans-Pacific transport of pollutants to the west coast of North America (Bailey et al. 2000; Jaffe et al. 2003; Wilkening et al. 2000). Fish contaminant studies in interior BC (Macdonald et al. 1999) and in lakes in the Yukon (Kidd et al. 1998; Kidd et al. 1995) showed levels which, in one case, necessitated fishery closure. Although significant residues in snow (Blais et al. 1998) and vegetation (CCME 1999, Davidson et al. 2003) have been measured in southwestern BC, no consistent survey of levels in fish has been reported. This survey addressed this significant data gap, and explored the extent and significance of POPs residues in freshwater fish tissues SW British Columbia. Preliminary results are summarized and discussed here, with a full presentation to be published elsewhere (Shaw and Gray, in prep.).

Method Summary
A total of 5-6 individuals of one dominant salmonid species (cutthroat trout, rainbow trout and Dolly Varden charr) from each of 17 lakes and reservoirs in SW BC were collected (Figure XX). Sites covered a wide range of physiographic conditions (elevation (0-1435m), watershed basin area/character, bathymetry).

Skin-off muscle fillets were analyzed for 84 PCB congeners, 26 organochlorine pesticides and dominant metabolites, total toxaphene and a suite of 10 chlorobornanes by GC/MS. In addition, sample extracts for PCB congener analysis were subsequently analyzed for 41 polybrominated diphenyl ether (PBDE) congeners.

Basic basin information, including hypsometry and land use/cover were derived by GIS.

Results Summary
Of the 120 analytes measured in this study, only 7 were not detected in any sample. Levels of all contaminants in muscle tended to be low, and with few exceptions did not exceed relevant Health and Welfare Canada residue limits consumption by humans or Council of Canadian Ministers of the Environment (CCME) guidelines for protection of wildlife consumers (CCME 1999).

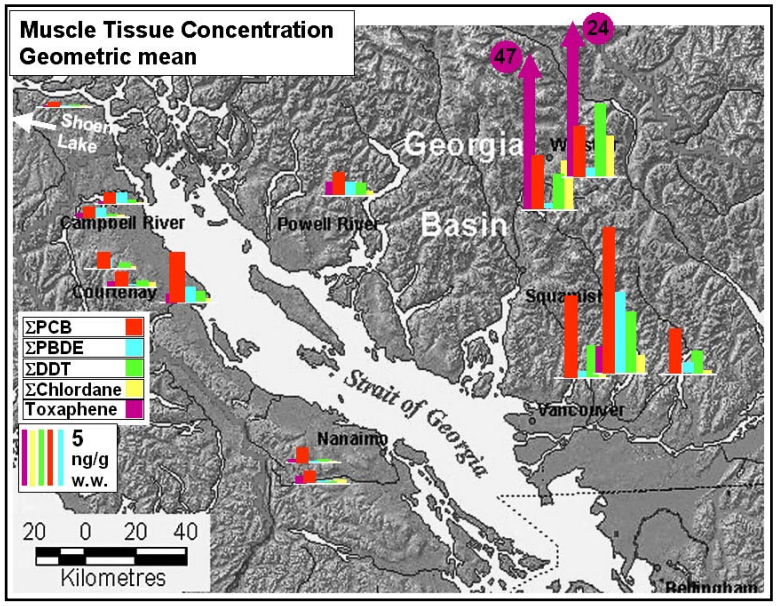

Figure 1. Contaminant summaries for rainbow and cutthroat trout muscle collected from lakes and reservoirs in south-west British Columbia. Bars represent geometric means of 3-6 individual analyses, in ng/g wet tissue weight.

Raw tissue concentrations varied widely both within and between sites, and the following is a summary of geometric means of the values from all sites. Geometric mean PCBs (sum of 84 congeners) ranged from 0.40 to 13.33 ng/g w.w. Mean ΣDDT (sum of *p,p'*-DDE, *o,p'*-DDE, *p,p'*-DDD, *o,p'*-DDD, *p,p'*-DDT, *o,p'*-DDT) ranged from 0.19 to 7.93 ng/g w.w., while ΣChlordane (sum of *cis* and *trans*-chlordane, *cis* and *trans*-nonachlor, oxychlordane, heptachlor and hepta-chlor epoxide) ranged from just 0.12 to 4.48 ng/g w.w. The ΣPBDE levels were similarly low, with concentrations for all but two sites ranging from 0.06 to 1.96 ng/g w.w. For the two remaining sites, average ΣPBDE levels were 6.35 and 6.90 ng/g w.w.

The spatial pattern of raw organohalogen concentrations in trout tissues is presented in Figure 1. Levels for most analytes were lowest in lakes on Vancouver Island, with the exception of somewhat elevated ΣPCB (geo. mean. 8.1 ng/g w.w.) in Comox Lake, near Courtenay, probably related to a local hydro-electric facility. Lakes in mountains immediately north of Vancouver tended to show elevated levels of contaminants which might be expected from their proximity to a large urban centre – these were the SPCB (4.0 – 13.3 ng/g w.w. geo mean) and, in one location, Coquitlam Lake with geo-mean ΣPDBE level of 6.1 ng/g w.w. The most remote lakes were within Garibaldi Provincial Park. Garibaldi Lake (elev. 1438m) and Cheakamus Lake (elev. 840m). Levels of ΣChlordane and ΣDDT were similar to the less isolated sites nearer Vancouver, but the highest toxaphene levels (to 180 ng/g w.w. in a single individual from Garibaldi Lake) of the study were found in these lakes.

Sources for the measured contaminants are varied, but the delivery mechanism to the waterbodies is due to atmospheric deposition. Strong correlations between the various analyte groups suggests there is a regional "signal" which has not been altered by lake-side sources such as a spill. Lakes in the vicinity of Vancouver and downwind of industries in Puget Sound showed elevated ΣPCBs and elevated ΣDDT at some sites, a probable consequence of release from historical use.

Elevated levels of both ΣChlordane, and, in particular ΣToxaphene in the alpine lakes of Garibaldi Park are perhaps a consequence of release of atmospherically transported contaminants from both snowpack and glacier ice. Two lines of evidence support this possibility. First is the pattern of the DDE/ΣDDT ratio amongst lakes. Earlier work has shown (Sanchez et al. 1993) that a relatively "fresh", un-degraded DDT is characterized by a relatively high proportion of DDE, which is the final and most stable metabolic product. Values are considerably lower in the two alpine lakes, both of

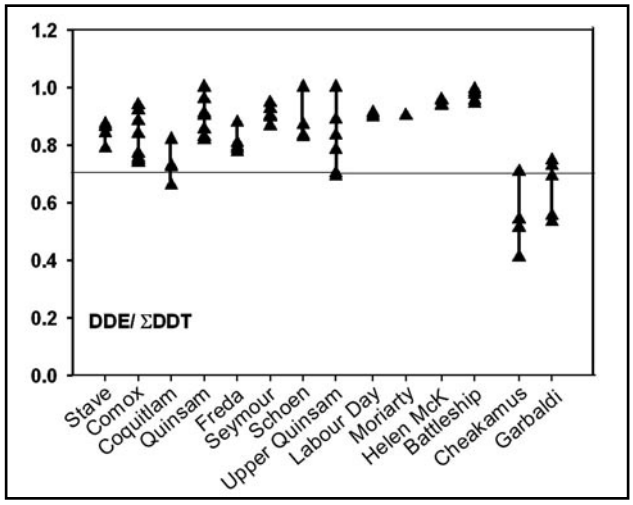

Figure 2. Plot of DDE/ΣDDT (sum of *p,p'*-DDE, *o,p'*-DDE, *p,p'*-DDD, *p,p'*-DDT, *o,p'*-DDT) for individual rainbow and cutthroat muscle tissues from lakes in southwestern British Columbia. The two lakes having values <0.7, Cheakamus and Garibaldi) are within Garibaldi Provincial Park and have predominantly glacial drainage.

which have greater than 20% of their total basin areas covered by glacial ice (Figure 2). The second line of evidence is an extremely poor relationship between both ΣChlordane and ΣToxaphene and lake elevation (linear regression, $p>0.5$), but an excellent relationship between the presence of glacial ice within the drainage basin and both ΣToxaphene and ΣChlordane (logistic regression, $p<0.05$). Observation of glacial contribution to organochlorine levels in lakes in Garibaldi Park corroborates previous work in the Canadian Rocky Mountains, where glacial ice has been shown to be both a significant reservoir of contaminants (Donald et al. 1999) and an important source of contaminants in lakes receiving glacial melt (Blais et al. 2001a, Blais et al. 2001b).

Contaminant levels in fish tissues measured in this work are probably, though not conclusively, a combination of three potential sources. A background level of contamination may be contributed from air-borne contaminants carried in westerly flows off the Pacific Ocean which could be contaminated over eurasia or through from the sea as the air masses move over the ocean (Jantunen and Bidleman 1996, Jantunen and Bidleman 1998., Wania and Mackay 1996). Local sources, such as industrial complexes and volatilization of residues from historical uses, contribute and produce recognizable spatial patterns in peak organochlorine levels. In some particular environments, such as the high-elevation lakes with significant glacial contribution to the total annual water budget, archived contaminants released from melting ice will be an important point-source of organochlorines (and other contaminants) for some time to come.

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